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54 Improved method for removing volatile substances from polyphenylene ether or polyphenylene ether/styrene resin compositions.

57 Volatile substances in polyphenylene ether or polyphenylene ether/polystyrene compositions are substantially reduced by extruding the resins in a single pass using a plurality of stages comprising water injection and vacuum venting.

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IMPROVED METHOD FOR REMOVING VOLATILE SUBSTANCES FROM POLYPHENYLENE ETHER OR POLYPHENYLENE ETHER/STYRENE RESIN COMPOSITIONS

This invention relates to the substantial removal of volatile substances from polyphenylene ether or polyphenylene ether/polystyrene compositions and, more particularly, to the reduction of volatile substances in an extruder comprising multiple stages of water injection followed by vacuum venting.

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BACKGROUND OF THE INVENTION

Polyphenylene ethers are a class of polymers which are widely used in industry, especially as engineering plastics in applications which require such properties as toughness and heat resistance. For many such applications polyphenylene ether resins are blended with various types of polystyrene resins to improve their processability. Recently it has become necessary to produce such compositions which are both substantially odorless and tasteless for food contact applications. It is therefore necessary that the polyphenylene ether or polyphenylene ether/polystyrene composition be substantially free from any volatile materials which have undesirable odors or would otherwise harm the food. Materials of this kind which are known to be present in polyphenylene ether or polyphenylene ether/polystyrene resins include toluene, dialkylamines such as di-n-butylamine, which are components of the catalyst used in preparing polyphenylene ether resins; styrene monomers, from degradation of any styrene resin and other by-products resulting from polyphenylene ether resin synthesis. In the case of poly(2,6-dimethyl-1,4-phenylene ether); 2,4, 6-trimethylanisole, 7-methyldihydrobenzofuran, 2,3-dihydrobenzofuran, 2,6-dimethylcyclohexanone and 2-ethylhex-2-enal are commonly present. Removal of sources of volatile odoriferous amines and oxygenated species is especially important because they are detectable in human organoleptic tests even in very low quantities.

Methods are known, to those skilled in the art, for removing volatile substances from polyphenylene ether or polyphenylene ether/polystyrene resins during vented extrusion, with or without the injection of water into the melt. For example, Kasahara et al., U.S. 4,369,278 extrudes polyphenylene ether and rubber reinforced polystyrene in a single pass, single stage vacuum vented extruder with optional water injection; Newmark, U.S. 3,633,880, extrudes plastic materials, which could include polyphenylene ether, in a single pass through an extruder with multiple vents and employs a specially designed screw to compress and decompress the melt aiding devolatilization without employing water. Commonly assigned pending U.S. Patent Application, Serial No. 156,046 filed February 16, 1988 (Attorney's Docket RD-18,017), extrudes polyphenylene ether in a single pass single stage system using up to 15 percent water and vacuum venting. Although all three of the above-mentioned methods reduce the amount of volatile substances in the resin, none of them reduce the amount of volatile odoriferous species down to a level where such materials are hardly detectable by analytical methods thus providing that articles made from them are substantially odorless, especially in food packaging applications.

Commonly assigned U.S. patent application of Banevicius, Serial No. , filed herewith, attorney's docket 335-2134 (8CN-8363), discloses a method for producing polyphenylene ether resin with a reduced content of phenolic by-products of the polymer synthesis, such as 2,4,6-trimethylanisole (2,4,6-TMA) and 7-methyldihydrobenzofuran (7-MDBF) in the initial production stage, but not the reduction of sources of residual amine components during post-production extrusion stages.

Commonly assigned U.S. Patent application of Banevicius, Serial No. , filed herewith, attorney's docket 335-2135 (8CN-8352/83) discloses a method of producing low odor polyphenylene ether/polystyrene resins by solution blending and employing a sequence of devolatilizers to reduce the content odoriferous impurities present in the resin.

It has now been discovered that if the prior art processes are modified by using a single pass through an extruder and dividing the water injection/vacuum venting stage into at least two stages, each consisting of at least one water injection step and at least one vacuum venting step, remarkable and highly unexpected reductions in volatile content and in sources of odoriferous amine content are achieved. By way of illustration, in comparison with a single pass, single stage extrusion, a polyphenylene ether composition containing 8.5 ppm of 2,4,6-trimethylanisole (TMA) is reduced to 2.6 ppm, but in a single pass, two stages, the TMA content is reduced to levels barely detectable by gas chromatography (0.27 ppm). If a 50:50 blend of polyphenylene ether and polystyrene is sent through an extruder in a single pass, single stage with 3 percent water injection and vacuum venting, the styrene content is reduced to 1130 ppm and the toluene

content is reduced to 271 parts per million; a back-to-back repetition using a single pass but two water injection/vacuum venting stages, using 1.5% by weight of water in each such stage, reduced the styrene content to 694 ppm and toluene content to 84 parts per million.

Furthermore, it has been surprisingly discovered that the use of a polyphenylene ether resin essentially free of phenolic by-products in the improved process of the present invention produces a resin with a very low content of any odoriferous components. This novel low odor resin can then be shaped into pellets for further processing or directly into solid sheets, molded or extruded articles, and foams which are highly desirable for use in food contact applications.

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DESCRIPTION OF THE DRAWING

The drawing is illustrative of the apparatus which may be used to carry out the present invention. It illustrates a vertical cross section of a twin screw extruder having two stages of water injection each of which has its own vacuum venting stages.

SUMMARY OF THE INVENTION

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According to the present invention there is provided an improved process for the reduction of volatile substances in a composition comprising

- (a) a polyphenylene ether resin, alone, or in combination with
- (b) a styrene resin, said composition comprising impurities selected from styrene monomer, toluene, sources of volatile odoriferous amines, volatile odoriferous oxygenated species, mixtures of any of them and the like, said process comprising extruding said composition at a temperature above its melting point in one pass in at least two stages, each said stage comprising water injection followed by vacuum venting, the total amount of water being divided between said stages and comprising up to about 15 percent by weight of said composition whereby the devolatilization efficiency of both toluene and trimethylanisole is greater than about 70 percent in comparison with the reduction obtained with the same composition in one pass in a single stage employing the same amount of water.

The water can, for example, comprise liquid water or steam. Carbon dioxide can also be used instead of water. Preferably the polyphenylene ether resin comprises poly(2,6-dimethyl-1,4-phenylene ether), poly(2,6-dimethyl-co-2,3,6-trimethyl-1,4-phenylene ether) or a mixture thereof, but may also comprise a functionalized polyphenylene ether wherein the functionalizing agent comprises fumaric acid, maleic anhydride, citric acid, trimellitic anhydride acid chloride or a mixture of any of the foregoing. Preferably the styrene resin comprises a polystyrene homopolymer or a rubber modified polystyrene.

Nucleating agents selected from an organic or inorganic particulate material or a mixture thereof may be added in an effective amount, preferably talc up to about 2 percent by weight of the composition. This appears to favorably induce bubble formation and thereby increase surface area during devolatilization.

Preferably the total amount of water to be injected into the extrudate is from about 1 to about 10 percent by weight of said composition, divided equally or unequally between the stages, and the pressure of the vacuum vents is set such that vapor velocity through the vent is kept below about 5-6 meters sec⁻¹.

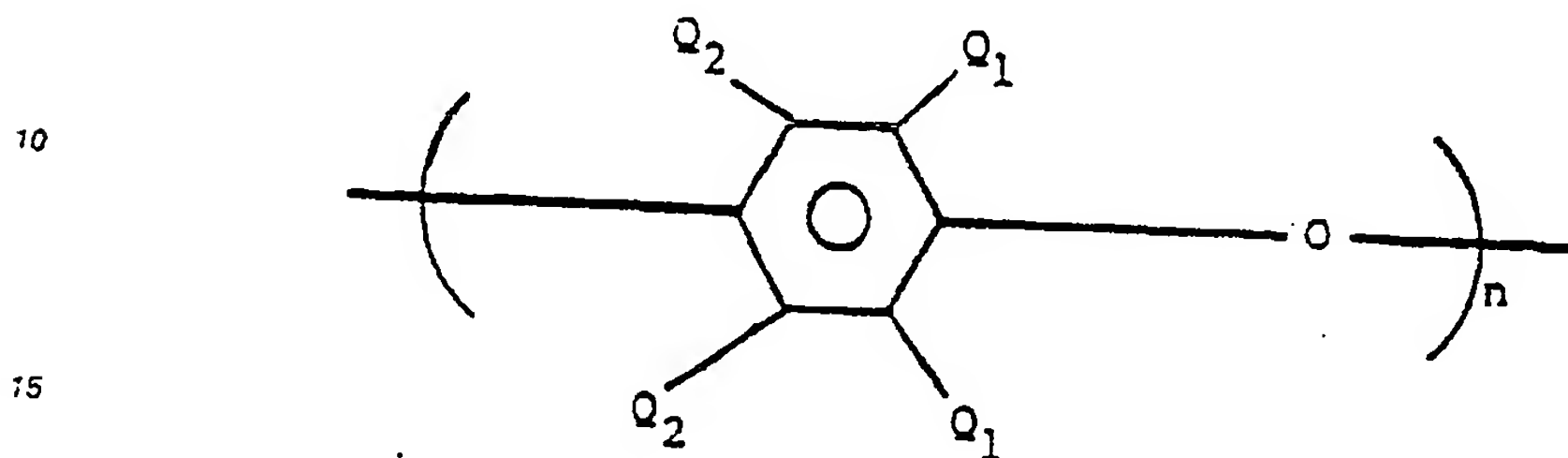
Preferred embodiments of the present invention also provide a process wherein the polyphenylene ether resin comprises a low odor polyphenylene ether resin comprising a 2,4,6-trimethylanisole content of less than about 50 parts per million by weight based on said resin and a source of amine content of greater than about 10,000 parts per million based on said resin. Preferably the low odor polyphenylene ether resin is prepared by oxidatively coupling a phenol in the presence of a catalyst in a solvent comprising a normally liquid aromatic hydrocarbon in a polymerization zone until formation of said resin and by-products are substantially complete and recovering said resin by the addition of a C₁ to C₆ alcohol to the polymer solution and recovering said aromatic hydrocarbon containing said by-products and distilling to reduce the content of said by-products and thereafter recycling said aromatic hydrocarbon essentially free of said by-products to the polymerization zone.

Among the preferred features of the present invention comprise processes for shaping the low odor composition into pellets, foamed boards or sheets, solid sheets, molded or extruded articles and the like.

DETAILED DESCRIPTION OF THE INVENTION

The polyphenylene ethers (also known as polyphenylene oxides) used as component (a) in the present invention are a well known class of polymers and are disclosed in Hay, U.S. 3,306,874 and 3,306,875.

The polyphenylene ethers favored for use in the practice of this invention generally contain structural units of the following formula



in which, independently, each Q₁ is hydrogen, halogen, primary or secondary lower alkyl containing up to 7 carbon atoms, phenyl, haloalkyl or amino alkyl wherein at least two carbon atoms separate the halogen or nitrogen atom from the benzene ring, hydrocarbonoxy, or haloalkylhydrocarbonoxy wherein at least two carbon atoms separate the halogen and oxygen atoms; and each Q₂ is independently hydrogen, halogen, primary or secondary lower alkyl, phenyl, haloalkyl, hydrocarbonoxy or haloalkylhydrocarbonoxy as defined in Q₁. Examples of suitable primary lower alkyl groups are methyl, ethyl, n-propyl, n-butyl, isobutyl, n-amyl, isoamyl, 2-methylbutyl, n-hexyl, 2,3-dimethylbutyl, 2-, 3- or 4-methylpentyl and the corresponding heptyl groups. Examples of secondary lower alkyl groups are isopropyl, sec-butyl and 3-pentyl. Preferably, any alkyl radicals are straight chain rather than branched. Most often each Q₁ is alkyl or phenyl, especially C₁₋₄ alkyl, and each Q₂ is hydrogen.

Both homopolymers and copolymers are included in the foregoing description. Suitable homopolymers are those containing, for example, 2,6-dimethyl-1,4-phenylene ether units. Suitable copolymers include random copolymers containing such units in combination with, for example, 2,3,6-trimethyl-1,4-phenylene ether units. Many suitable random copolymers, as well as homopolymers, are disclosed in the patent literature including the various Hay patents. Also contemplated are graft copolymers, including those prepared by grafting onto the polyphenylene ether chain such vinyl monomers as acrylonitrile and vinyl aromatic compounds (for example, styrene) and such polymers as polystyrenes and elastomers. Still other suitable polyphenylene ethers are the coupled polyphenylene ethers in which the coupling agent is reacted with the hydroxy groups of the two polyphenylene ether chains to increase the molecular weight of the polymer. Illustrative of the coupling agents are low molecular weight polycarbonates, quinones, heterocycles and formals.

The polyphenylene ether generally has a molecular weight (number average, as determined by gel permeation chromatography, whenever used herein) within the range of about 5,000 to 40,000. The intrinsic viscosity of the polymer is usually in the range of about 0.30 to 0.6 deciliters per gram (dl/g) as measured in solution in chloroform at 25° C.

The polyphenylene ethers are typically prepared by the oxidative coupling of at least one corresponding monohydroxyaromatic compound. Particularly useful compounds are 2,6-xlenol (wherein each Q₁ is methyl and each Q₂ is hydrogen), whereupon the polymer may be characterized as a poly(2,6-dimethyl-1,4-phenylene ether), and 2,3,6-trimethylphenol (wherein each Q₁ and one Q₂ is methyl and the other Q₂ is hydrogen).

A variety of catalyst systems are known for the preparation of polyphenylene ethers by oxidative coupling. There is no particular limitation as to catalyst choice and any of the known catalysts can be used. For the most part, they contain at least one heavy metal compound such as a copper, manganese or cobalt compound, usually in combination with various other materials.

A first class of preferred catalyst systems consists of those containing a copper compound. Such catalysts are disclosed, for example, in U.S. Patents 3,306,874, 3,306,875, 3,914,266 and 4,028,341. They are usually combinations of cuprous or cupric ions, halide (i.e., chloride, bromide or iodide) ions and at least one amine.

Catalyst systems containing manganese compounds constitute a second preferred class. They are generally alkaline systems in which divalent manganese is combined with such anions as halide, alkoxide or phenoxide. Most often, the manganese is present as a complex with one or more complexing and/or

chelating agents such as dialkylamines, alkanolamines, alkylenediamines, omega-hydroxyaromatic aldehydes, o-hydroxyazo compounds, beta-hydroxyoximes (monomeric and polymeric), o-hydroxyaryl oximes and beta-diketones. Also useful are known cobalt-containing catalyst systems. Suitable manganese and cobalt-containing catalyst systems for polyphenylene ether preparation are known in the art by reason of disclosure in numerous patents and publications.

The process of this invention may also be employed with functionalized polyphenylene ethers. These may be prepared by the reaction of at least one functionalizing agent with a polyphenylene ether. The functionality of the functionalized polyphenylene ether may be present on the end group; for example, as a result of a reaction with the phenolic terminal hydroxy group. Functionalization may also involve one of the aromatic groups in the aromatic rings in the polymer chain, or an alkyl group attached thereto.

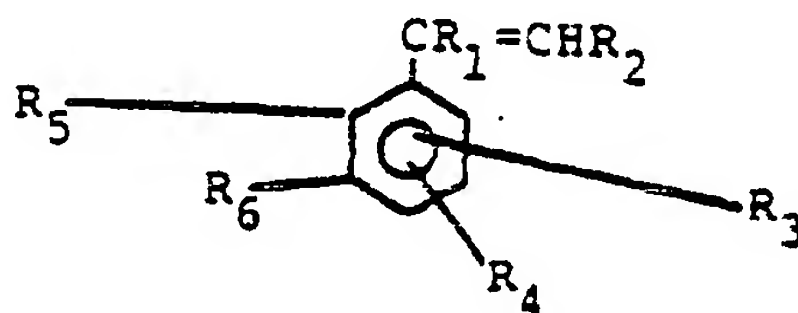
One method of functionalizing the polyphenylene ether is by reaction with at least one compound containing (a) a carbon-carbon double or triple bond, hydroxy group, alkoxy group, arloxy group or acyl halide group, and also (b) a carboxylic acid, acid salt, acid anhydride, acid amide, acid ester or imido group. A wide variety of such compounds are suitable for use in the invention. Many illustrative compounds are listed in U.S. 4,315,086 and U.S. patent application Serial No. 885,497 filed July 14, 1986. They include maleic, fumaric, itaconic and citraconic acids and their derivatives, various unsaturated fatty oils and the acids derived therefrom, relatively low molecular weight olefinic acids such as acrylic acid and its homologs, and the like.

Other contemplated functionalizing agents are the aliphatic polycarboxylic acids and derivatives thereof disclosed in U.S. patent application Serial No. 736,489 filed May 20, 1985. Illustrative polycarboxylic acids of this type are citric acid, maleic acid and agaric acid and their esters, amides and salts.

Still another class of contemplated functionalizing agents are disclosed in U.S. patent 4,600,741. Illustrative compounds within this class are carboxymethylsuccinic anhydride acid chloride and trimellitic anhydride acid chloride (TAAC).

The present invention also includes resin compositions comprising polystyrene resins in addition to the polyphenylene ether resin. Polystyrene resins are generally added to the polyphenylene ether resin in order to improve the processability of the resin.

The polystyrene resins are broadly defined herein and based at least in part from compounds of the formula



wherein R_1 and R_2 are selected from the group consisting of lower alkyl or alkenyl groups of from 1 to 6 carbon atoms and hydrogen; R_3 and R_4 are selected from a group consisting of chloro, bromo, hydrogen and lower alkyl groups of from 1 to 6 carbon atoms, R_5 and R_6 are selected from a group consisting of hydrogen and lower alkyl and alkenyl groups of from 1 to 6 carbon atoms or R_5 and R_6 may be concatenated together with hydrocarbonyl groups to form a naphthyl group.

Compounds within the above formula include styrene and its homologs and analogs. In addition to styrene, examples include alpha-methyl styrene, paramethyl styrene, 2,4-dimethyl styrene, chlorostyrene, dichlorostyrene, bromostyrene, dibromostyrene, p-tert-butyl styrene, p-ethylstyrene, vinylxylene, divinylbenzene and vinylnaphthalene. Styrene is especially preferred.

Also contemplated for use in the present invention are rubber modified polystyrenes, including high impact polystyrenes generally known as HIPS. In general, these modified polystyrene resins are made by adding rubber or rubber precursors, such as dienes, polydienes, olefin rubbers, acrylonitrile rubbers, acrylic rubbers and the like, during or after polymerization of the styrene, to yield an interpolymer of rubber and polystyrene, a physical admixture of rubber and polystyrene, or both, depending on the particular process employed.

Suitable rubber modifiers include polybutadiene, polyisoprene, polychloroprene, ethylene-propylene copolymers (EPR), ethylene-propylene-diene (EPD) rubbers styrene-butadiene copolymers (SBR), polyacrylates, polynitriles, mixtures thereof and the like. The amount of rubber employed will vary, depending on such factors as the process of manufacture and individual requirements.

Included within this family of materials for purposes of this invention are more recently developed forms

in which such factors as the rubber particle size, the gel and cis contents of the rubber phase, and the rubber volume percent are regulated or controlled to achieve improvements in impact resistance and other properties. These kinds of rubber modified polystyrenes and HIPS are described in the patent literature, including Katchman and Lee, U.S. 4,128,602 and Cooper and Katchman, U.S. 4,528,327.

Also contemplated as suitable for use herein are rubber modified polystyrenes and HIPS having morphological forms which are sometimes referred to as core-shell, comprising particles of rubber encapsulated polystyrene dispersed in a matrix of polystyrene resin. Examples of this type are disclosed in Bennett and Lee, U.S. 4,513,120 as well as the above-mentioned U.S. 4,528,327.

Polyphenylene ether (PPE) resins and polystyrene (PS) resins are combinable in all proportions, e.g., from about 1 to about 99 parts by weight polyphenylene ether and from about 99 to about 1 part by weight polystyrene. It is contemplated, however, that low density compositions of the present invention are comprised of at least 2 weight percent PPE, based upon the weight of PPE and PS taken together. Compositions containing less than 2 weight PPE are considered to be primarily polystyrene compositions and do not generally exhibit the preferred property improvements associated with PPE/PS blends. It is well known that the addition of PPE to polystyrene resins offers improvements in impact strength, flammability ratings, tensile strength and other mechanical properties. Conversely, polystyrene is typically blended with polyphenylene ether resins to offer better processability for many thermoplastic processes.

Typical PPE/PS blends useful in the practice of the present invention will be comprised of between 5 to 95 percent and preferably 20 to 80 percent by weight PPE and 95 to 5 percent and preferably 80 to 20 percent by weight PS based upon the weight of the two resins taken together.

It is also contemplated in the present invention to add an effective amount of an organic or inorganic particulate material as a nucleating agent to the resin composition to induce bubble formation and increase surface area during devolatilization. One can use ground glass, carbon black, talc, and the like. Preferably the nucleating agent comprises talc in an amount of up to about 2 percent by weight based on said resin composition.

An essential step in the method of this invention is extrusion of the polyphenylene ether composition with styrene resin. Extrusion may be conducted using any known equipment for this operation, including single-screw and twin-screw extruders. See, for example, *Modern Plastics Encyclopedia/88*, October 1987, McGraw Hill, New York, p. 228 - 238. Especially suitable for amine removal is a co-rotating twin screw extruder.

Since multiple stages of water injection and vacuum venting are also essential features of this invention, the presence of suitable ports in the extruder for injection of a stripping agent and vents for vacuum venting are mandatory. The stripping agent may be introduced at any point upstream of the first vent or set of vents; however, it is highly preferred to introduce the stripping agent through a port located at a point in the extruder where the polymer composition has been converted into a fully developed polymer, since this facilitates intimate contact with the polymer.

Water or steam are the preferred stripping agents, and the proportion employed is up to about 15 percent by weight of the polymer composition, to be divided equally, or unequally, among the two or more injection ports located along the length of the extruder barrel. The preferred proportion is about 1 - 10 percent, since an amount within this range is generally optimally effective for removal of volatiles without burdening the vacuum system. For example, where two injection ports are present and about 5 percent total water by weight of polymer composition is to be used, each injection port would inject about 2½ percent of the water into the extrudate.

The degree of vacuum will depend on several factors, including the proportion of volatile impurities in the polyphenylene ether or polyphenylene ether/poly styrene resin and the amount of water to be employed. In general, it is preferable to limit the vapor velocity across the vent interface to about 5-6 meters sec⁻¹ and the pressure should be set accordingly.

In most instances, maximum or near maximum rate of rotation of the extruder screw or screws should be maintained for effective volatilization. The rotation rate will to some extent be dependent on the equipment used, but values in the range of about 300 to about 500 revolutions per minute are generally sufficient.

Referring to the drawing, extruder 1 comprises a heated barrel 2 and a multi-flight screw 4 adapted to co-rotate with a twin screw (not shown). Feed hopper 6 located at the upstream end is adapted to receive polyphenylene ether or polyphenylene ether and polystyrene and any conventional additives and, if desired, a nucleating agent, such as talc. As the resin moves downstream, it is heated and melted. The resin melt then encounters stage 20 comprising water injection port 8 and vacuum vents 10 and 12. The resin melt which exits stage 20 immediately encounters the second stage 22 comprising water injection port 14 and vacuum vents 16 and 18. Finally, the material exits the extruder at front barrel outlet 24. The exiting product

can be extruded into shapes or cut into pellets for further processing in accordance with conventional techniques.

It is further contemplated to employ a polyphenylene ether resin essentially free from phenolic odoriferous by-products of polymer synthesis, such as 2,4,6-trimethylanisole and 7-methyldihydrobenzofuran, removed from the normally liquid aromatic hydrocarbon recycle stream by distillation. The distillation may be performed in any suitable distillation column. The recycle aromatic hydrocarbon stream is fed into a column shell comprising various plates or trays, which are used to bring the vapor and liquid phases of the feed material into intimate contact, stacked one above the other inside the enclosed column. Optionally, a packed column may be employed. The aromatic hydrocarbon solution is boiled and the purified aromatic hydrocarbon vapor is collected and condensed at the top of the column, while the impurities remain liquid and are collected at the bottom of the column. The number of trays necessary is dependent upon the degree of purity desired in the distillate and the type of tray used. Suitable for the practice of this invention are distillation columns employing sieve type trays ranging from about 40 to about 65 in number.

Distillation is carried out to render the recycle aromatic hydrocarbon essentially free of by-product content. Essentially free is defined as the level of odoriferous impurities present in the recycle aromatic hydrocarbon such that recycling the aromatic hydrocarbon to the polymerization zone results in a very low odor product resin, not detectable by individuals with highly developed olfactory systems. Preferably, the 2,4,6-trimethylanisole content in the recycle aromatic hydrocarbon is reduced to less than about 5 parts per million and the 7-methyldihydrobenzofuran is reduced to less than about 50 parts per million.

Other embodiments of the process of the present invention, include shaping the low odor composition into pellets, solid sheets, foamed sheets or boards, molded or extruded articles and the like.

Any conventional hot or cold pelletizing or dicing systems may be used to form pellets. Cold cutting systems are generally dicing, strand pelletizing and strand (forced conveyance) pelletizing systems. Hot cutting systems generally comprise water ring pelletizers, hot face pelletizers, underwater pelletizers and centrifuged pelletizers. See generally Modern Plastics Encyclopedia/88, McGraw-Hill, 1987, pp. 340-342. Solid sheets are generally formed by extending the molten composition through dies specially suited for forming solid sheets, such as flat sheet dies, although any die which will produce a solid sheet may be employed. See generally, Modern Plastics Encyclopedia/88, McGraw Hill, 1987, pp. 236-237. Extruded or molded articles may be produced in any conventional process and apparatus known to those skilled in the art.

Optionally, the low odor composition may be foam processed. Any suitable apparatus for extruding foamed sheets or boards may be employed in this shaping process. See for example, Modern Plastics Encyclopedia/88, McGraw Hill 1987, pp 238-246. Especially suitable for the practice of the present invention are tandem extruders. The resin composition is fed into a first mixing-melting single or twin screw type extruder and melted and mixed therein with a liquid blowing agent at sufficient temperature and shear to provide an intimate blend.

During the blending step it is also contemplated to introduce conventional additives into the polymer composition melt. These include nucleating agents, flame retardants, thermal and color stabilizers, antioxidants, processing aids, plasticizers, reinforcing and extending fillers, pigments, dyes and the like. Each of these may be utilized to a greater or lesser degree depending on the desired final properties of the foamed product. Conventional surfactants and nucleants may also be utilized, such as zinc or tin stearates, maleates and fumarates.

Suitable nucleating agents, which aid in controlling the foam cell size and number of foam cells, usually comprise a fine powder such as talc or a combination of citric acid and bicarbonate of soda.

Suitable blowing agents to be used in the melt produced in the extruder include conventional hydrocarbons, chlorofluorocarbons, and hydrochlorofluorocarbons. Hydrocarbon blowing agents will include aliphatic hydrocarbons, especially those having 4 to 7 carbon atoms such as pentane, isopentane, pentene, hexane, heptane, butane and the like. Chlorofluorocarbon blowing agents include CCl_3F , CCl_2F_2 , $\text{C}_2\text{Cl}_3\text{F}_3$, C_2ClF_5 , CHClF_2 and $\text{CCl}_2\text{F}_2\text{-CClF}_2$. These are commercially available as Freon® 11, Freon® 12, Freon® 22, Freon® 113, Freon® 115 and Freon® 114. Hydrochlorofluorocarbon blowing agents include compounds such as chlorodifluoromethane, dichlorofluoromethane, dichlorodifluoroethane and the like.

Although the extrudate can be foamed through a die in the first extruder, preferably the extrudate is transferred through a pressurized closed conduit to a second single or twin screw extruder.

The conduit should be heated to maintain melt consistency. In the second extruder, the melt is cooled and exits as a foam at a die located at the downstream end of the extruder barrel.

As an alternative, the blowing agent can also be introduced into the devolatilizing extruder to obtain a melt which contains the liquid blowing agent under pressure. This material can be foamed directly out a die

at the downstream end of the devolatilizing extruder or may be transferred to single or tandem foam extruders for foaming.

Also contemplated by the present invention is combining the low odor composition of the present invention with let-down resins, including polyphenylene ether resins, prior to further processing or shaping.
5 Resins such as a polystyrene, a rubber modified polystyrene, copolymers of styrene and acrylonitrile, a poly(butylene terephthalate), a poly(bisphenol-A carbonate), a poly(etherimide ester), a poly(ester carbonate), a polyamide resin, ABS resins, and the like, or a mixture of any of them can be used as let-down resins.

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DESCRIPTION OF PREFERRED EMBODIMENTS

The following examples illustrate the present invention. They are not to be construed to limit the claims
15 in any manner whatsoever.

EXAMPLE 1

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A poly(2,6-dimethyl-1,4-phenylene ether)/polystyrene homopolymer blend comprising about 50 parts by weight of each resin component is fed into an extruder equipped with a two stage water injection/vacuum venting system at a rate of 318 kg/hr, a screw speed of 350 rpm, barrel temperature setpoints at 343 °C, a melt temperature of 360 °C wherein water is injected at a rate of 1.5 weight percent of the feed at each
25 injection port and the vacuum pressure set at about 130 - 150 mbar. The starting material contains 9 ppm of TMA, 0.5 percent of DBA and 1500 ppm of toluene. A control sample is fed into a single stage water injection vacuum venting system extruder at the same processing parameters except that water is injected at 3 weight percent of the feed rate through the single injection port. The results of volatile substance removal are set forth below in Table 1.

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TABLE 1

VOLATILES REDUCTION 50/50 PPE/PS EXTRUDATE		
Examples	1*	1
EXTRUDER PARAMETERS		
Feed Rate, kg/hr	318	318
Screw Speed, rpm	350	350
Barrel Temp., °C	343	343
Melt Temp., °C	360	360
Water Injection Rate, %	3	1.5
Vacuum Pressure, mbar	130 - 150	130 - 150
EXTRUDATE VOLATILES		
Styrene monomer, ppm	1130	694
Toluene, ppm	271	84
DBA, %	0.18	0.15
TMA, ppm	2.6	0.27
7-MDBF	3.3	0.1

* Control Sample

DBA = di-butyl amine

TMA = 2,4,6-trimethylanisole

7-MDBF = 7-methyldihydrobenzofuran

The table clearly shows that using an equal amount of water in a two stage process results in a vastly greater reduction of volatile impurities from the extrudate than can be accomplished in a one stage process.

EXAMPLE 2

A resin composition comprising 70 parts by weight of poly(2,6-dimethyl-1,4-phenylene ether) and 30 parts by weight of polystyrene homopolymer is fed into an extruder equipped with a single stage water injection vacuum venting system at a rate of 81.8 kg/hr, a screw speed of 350 RPM, barrel temperature setpoints at 315 °C, wherein water is injected at a rate of 2.5% and 5% and the vacuum pressure is set at 35 mbar. The resin composition contains about 0.7% DBA by weight and also 175 ppm TMA by weight. A resin composition containing 20 ppm of TMA is extruded under the same conditions, except that water is injected at a rate of 2.5 weight % of the feed at each of two water injection ports. The results of volatile substance removal are set forth below in Table 2.

TABLE 2

EXTRUDER PARAMETERS	2A*	2B*	2C*	2
Feed rate, kg/hr	82	82	82	82
Screw speed, rpm	350	350	350	350
Barrel Temp., °C	315	315	315	315
Water injection rate, %	2.5	5	0	5
Number of injection stages	1	1	0	2
Vacuum pressure, mbar	35	35	35	70
FEEDSTOCK VOLATILES				
TMA, ppm	175	175	175	20
DBA, wt. %	0.7	0.7	0.7	0.7
% TMA removed	85	86	80	97
EXTRUDATE VOLATILES				
TMA, ppm	26	24	34	0.5
DBA, wt. %	0.32	0.31	0.37	0.17
% TMA removed	85	86	80	97

* = Control Sample

TMA = 2,4,6-trimethylanisole

DBA = di-butyl amine

wppm = weight parts per million

The table clearly shows using an equal amount of water in a two stage process results in a vastly greater reduction of volatile impurities from the extrudate than can be accomplished in a one-stage process.

EXAMPLE 3

The procedure of Example 2 is repeated for a resin composition comprising 49 parts by weight of poly(2,6-dimethyl-1,4-phenylene ether), 49 parts by weight of polystyrene homopolymer and 2 parts by weight of talc. Many tiny bubbles are created in the vacuum vented section. The extrudate is comminuted into pellets and the pellets are odorless. Without talc, occasional individuals with highly developed olfactory systems sometimes state that the pellets are slightly odorous. When molded into soup containers, the products of this example pass the soup taste test, a very discriminating test for low odor in food packaging.

The above-mentioned patents, applications and publications as well as test methods are incorporated herein by reference.

Many variations of the present invention will suggest themselves to those skilled in this art in light of the above detailed description. For example, instead of polyphenylene ether in Example 1, a mixture of 100 parts of polyphenylene ether and 0.7 parts of fumaric acid can be fed into the extruder and a functionalized polyphenylene ether with a very low odoriferous amine content can be obtained. Instead of poly(2,6-dimethyl-1,4-phenylene ether), a poly(2,6-dimethyl-co-2,3,6-trimethyl-1,4-phenylene ether) can be used. Additionally, a low odor polyphenylene ether resin may be employed. A three stage extrusion can be used in which the first stage comprises carbon dioxide injection followed by vacuum venting, followed by two water injection vacuum venting stages. Steam can be used instead of water. All such obvious variations are within the full intended scope of the appended claims.

Claims

1. A process for the reduction of volatile substances in a composition comprising (a) a polyphenylene ether resin, alone, or in combination with

- (b) a styrene resin, said composition comprising impurities selected from styrene monomer, toluene, volatile odoriferous oxygenated species, sources of volatile odoriferous amines, mixtures of any of them and the like, said process comprising extruding said composition at a temperature above its melting point in one pass in at least two stages, each said stage comprising water injection followed by vacuum venting, the total amount of water being divided between said stages and comprising up to about 15 percent by weight of said composition whereby the devolatilization efficiency of both toluene and trimethylanisole is greater than about 70 percent in comparison with the reduction obtained with the same composition in one pass in a single stage employing the same amount of water.
2. A process as defined in Claim 1 wherein said composition comprises polyphenylene ether, alone.
3. A process as defined in Claim 1 wherein said polyphenylene ether resin component (a) comprises poly(2,6-dimethyl-1,4-phenylene ether), poly(2,6-dimethyl-co-2,3,6-trimethyl-1,4-phenylene ether) or a mixture thereof.
4. A process as defined in Claim 1 wherein said styrene resin component (b) comprises a polystyrene homopolymer resin.
5. A process as defined in Claim 1 wherein said styrene resin component (b) comprises a rubber modified styrene polymer.
6. A process as defined in Claim 1 wherein said polyphenylene ether resin component (a) comprises a functionalized polyphenylene ether.
7. A process as defined in Claim 6 wherein said functionalizing agent is fumaric acid, maleic anhydride, citric acid, trimellitic anhydride acid chloride or a mixture of any of the foregoing.
8. A process as defined in Claim 7 wherein said functionalizing agent is maleic anhydride.
9. A process as defined in Claim 1 wherein the temperature in the extruder is above about 280 °C and the water injected comprises liquid water.
10. A process as defined in Claim 1 wherein the temperature in the extruder is above about 280 °C and the water injected comprises steam.
11. A process as defined in Claim 1 wherein the total amount of water injected is from about 1 to about 10 percent by weight of the polymer composition.
12. A process as defined in Claim 1 wherein the pressure during extrusion is in the range of about 550 to about 720 torr.
13. A process as defined in Claim 1 wherein an effective amount of a nucleating agent selected from an organic or an inorganic particulate material or mixture thereof is added to the composition to induce bubble formation and increase surface area during devolatilization.
14. A process as defined in Claim 13 wherein said nucleating agent comprises talc in an amount of up to about 2 percent by weight based on said composition.
15. A process as defined in Claim 1 including the step of injecting carbon dioxide into the extruder in at least one injection stage.
16. A process as defined in Claim 1 including setting the pressure of the vacuum vent such that the vapor velocity through the vent is kept below about 5-6 meters sec⁻¹.
17. A process as defined in Claim 1 wherein said polyphenylene ether resin component (a) comprises from about 5 to about 95 parts by weight and said polystyrene resin component (b) comprises from about 95 to about 5 parts by weight.
18. In a process for the reduction of volatile substances in a composition comprising
 (a) a polyphenylene ether resin, alone, or in combination with
 (b) a styrene resin, said composition comprising impurities selected from styrene monomer, toluene, volatile odoriferous oxygenated species, sources of volatile odoriferous amines, mixtures of any of them, and the like; said process comprising extruding said composition at a temperature above about 280 °C in one pass in the presence of water followed by vacuum venting, the improvement which comprises carrying out the process in at least two stages each said stage comprising water injection followed by vacuum venting, the total amount of water being divided between said stages and comprising up to about 15 percent by weight of said composition, whereby the devolatilization efficiency of both toluene and trimethylanisole is greater than about 70 percent in comparison with the reductions obtained with the same composition in one pass in a single stage employing the same amount of water.
19. A process as defined in Claim 1 wherein said polyphenylene ether resin comprises a low odor polyphenylene ether resin having a 2,4,6-trimethylanisole content of less than about 50 parts per million by weight based on said resin and sources of odoriferous amine content of greater than about 10,000 parts per million based on said resin.
20. A process as defined in Claim 19 wherein said low odor polyphenylene ether resin is prepared by oxidatively coupling a phenol in the presence of a catalyst in a solvent comprising a normally liquid

aromatic hydrocarbon in a polymerization zone until formation of said resin and by-products are substantially complete and recovering said resin by the addition of a C₁ to C₆ alcohol to the polymer solution and recovering said aromatic hydrocarbon containing said by-products and distilling to reduce the content of said by-products and thereafter recycling said aromatic hydrocarbon essentially free of said by-products to the polymerization zone.

21. A process as defined in Claim 20 wherein said phenol feed into the reactor comprises 2,6-xylenol.

22. A process as defined in Claim 20 wherein said normally liquid aromatic hydrocarbon comprises toluene.

23. A process as defined in Claim 20 wherein said distilling comprises distilling said aromatic hydrocarbon and said by-products to reduce the content of 2,4,6-trimethylanisole by-product present in said aromatic hydrocarbon to less than about 5 weight parts per million based on said aromatic hydrocarbon.

24. A process as defined in Claim 19 wherein said polyphenylene ether resin comprises a by-product 7-methyldihydrobenzofuran content of about 0.5 to about 50 weight parts per million based on said resin.

25. A process as defined in Claim 19 which also comprises step (c) comprising shaping the low odor composition.

26. A process as defined in Claim 25 wherein said step (c) comprises pelletizing said low odor composition.

27. A process as defined in Claim 25 wherein said step (c) comprises foaming said low odor composition.

28. A process as defined in Claim 27 wherein said step (c) comprises

(i) feeding said low odor composition into at least one extruder and mixing it with a blowing agent; and

(ii) foaming the mixture through a die to form a shaped, foamed sheet or board having very low odor in human organoleptic tests.

29. A process as defined in Claim 28 wherein said step (i) comprises feeding said composition into tandem extruders, the first extruder being adapted to melt and mix said composition and said blowing agents into the polymer melt and the second extruder is adapted for cooling the melt prior to foaming.

30. A process as defined in Claim 28 wherein said blowing agent comprises a hydrocarbon, a chlorofluorocarbon, a hydrochlorofluorocarbon or a mixture thereof.

31. A process as defined in Claim 30 wherein said blowing agent comprises chlorodifluoromethane.

32. A process as defined in Claim 28 which also comprises adding a nucleating agent in step (i) in an amount sufficient to aid in regulating the size of foam cell and number of foam cells.

33. A process as defined in Claim 25 wherein said step (c) comprises extruding said composition through a suitable die to form a solid sheet having very low odor in human organoleptic tests.

34. A process as defined in Claim 25 wherein said step (c) comprises

(i) adding said blowing agent directly into the extruder of step (b) to yield a low odor composition containing a liquid blowing agent under pressure; and

(ii) feeding said low odor composition into an extruder equipped with a foaming die; and

(iii) foaming said composition through said die to form a shaped foamed sheet or board having very low odor in human organoleptic tests.

35. A process as defined in Claim 25 wherein said step (c) comprises extruding said composition through a suitable die to form an extruded article having very low odor in human organoleptic tests.

36. A process as defined in Claim 25 wherein said step (c) comprises molding said composition to form a molded article having very low odor in human organoleptic tests.

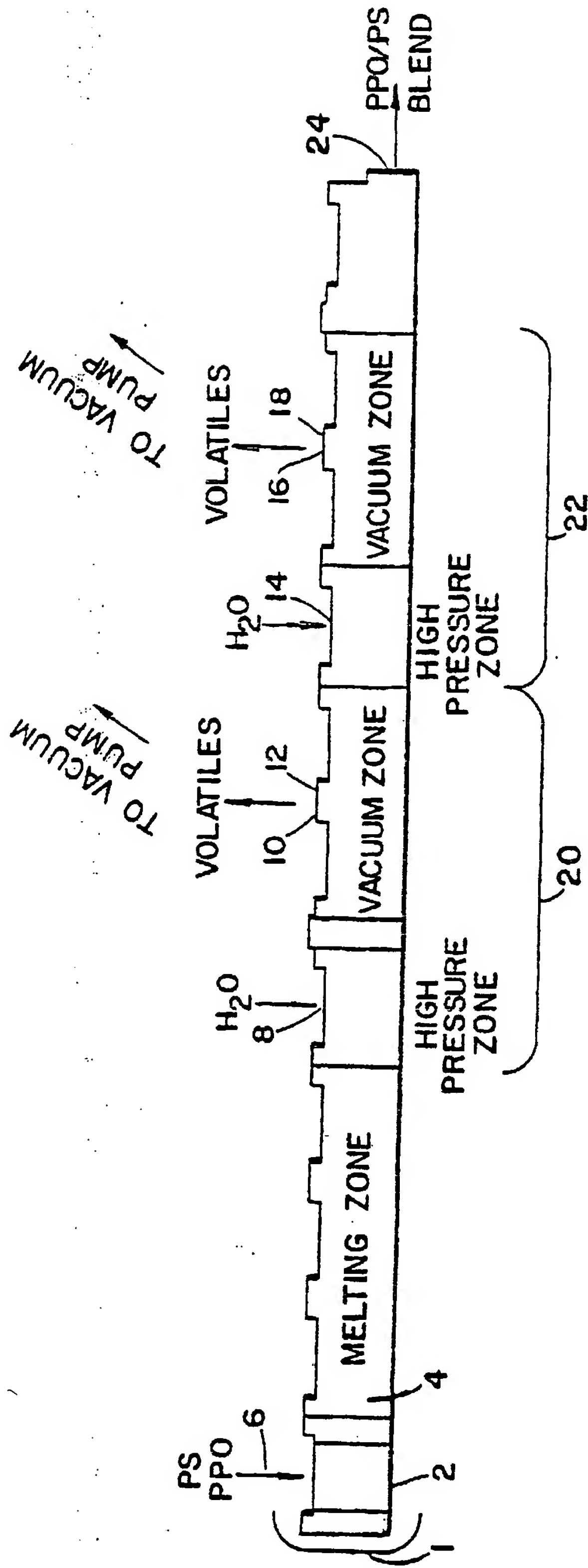
37. A low odor polyphenylene ether or polyphenylene ether/polystyrene pellet produced by the process as defined in Claim 26.

38. A low odor polyphenylene ether or polyphenylene ether/polystyrene foamed sheet or board produced by the process as defined in Claim 28.

39. A low odor polyphenylene ether or polyphenylene ether/polystyrene solid sheet produced by the process as defined in Claim 33.

40. A low odor polyphenylene ether or polyphenylene ether/polystyrene extruded article produced by the process as defined in Claim 35.

41. A low odor polyphenylene ether or polyphenylene ether/polystyrene molded article produced by the process as defined in Claim 36.





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Publication number:

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12

EUROPEAN PATENT APPLICATION

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54 Improved method for removing volatile substances from polyphenylene ether or polyphenylene ether/styrene resin compositions.

57 Volatile substances in polyphenylene ether or polyphenylene ether/polystyrene compositions are substantially reduced by extruding the resins in a single pass using a plurality of stages comprising water injection and vacuum venting.

EP 0 375 937 A3



European Patent
Office

EUROPEAN SEARCH REPORT

Application Number

EP 89 12 1608

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
X,Y	DE-A-3 603 798 (HULS) * Claims; column 3, lines 56-59; examples * ---	1-41	C 08 L 71/12 C 08 L 25/00
X	EP-A-0 071 093 (BASF) * Claims; page 7, lines 13-20 * ---	1-41	
P,Y	US-A-4 831 115 (J.C. GOLBA et al.) * Claims * -----	1-41	
			TECHNICAL FIELDS SEARCHED (Int. Cl.5)
			C 08 G C 08 L C 08 J
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 04-10-1990	Examiner DERAEDT G.
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ----- & : member of the same patent family, corresponding document			

EPO FORM 1503 03.82 (P0401)

XP-002303242

(C) WPI/Derwent

AN - 1988-342863 [48]

A - [001] 014 03- 034 036 037 040 055 056 058 13- 147 151 213 214 216 219
316 331 368 402 414 415 417 437 456 476 516 528 541 578 634 649 688

AP - JP19870091144 19870414

CPY - SUMO

DC - A14 A89 G06

DR - 0673-S 0708-S

FS - CPI

IC - B29B9/10 ; B29D11/00 ; B29K71/00 ; C08J3/12

KS - 0002 0003 0016 0218 0229 0297 0298 0304 0305 0318 0319 1279 1359 1361
2318 2327 2393 2394 2459 2510 2545 2587 2600 2676 2841 2851 3003 3310

MC - A04-C01 A05-H07 A07-A04E A11-A03 A11-C01 A12-L02A A12-L03 G06-D07

PA - (SUMO) SUMITOMO CHEM IND KK

PN - JP63256427 A 19881024 DW198848 006pp

PR - JP19870091144 19870414

XA - C1988-151765

XIC - B29B-009/10 ; B29D-011/00 ; B29K-071/00 ; C08J-003/12

AB - J63256427 Prodn. involves dissolving a resin compsn. which comprises a polymeric part made from aromatic vinyl monomer and a poly(phenylene ether) part, into an organic solvent, filtering the soln. to remove fine particulate foreign matter, supplying the soln. to a vent type screw extrusion granulation to remove the organic solvent, and melt-moulding the resin compsn.

- Pref. aromatic vinyl monomer is styrene, alpha-methylstyrene, etc. The polyphenylene ether is poly (2,6-dimethyl -1,4-phenylene) ether, poly (2-methyl-6-ethyl 1,4-phenylene) ether, etc. The organic solvent is benzene, toluene, etc.

- USE/ADVANTAGE - For optical disk substrates for recording or reproducing optical information, optical cards, lenses, prisms, etc. The filtration reduces content of particulate foreign matter. The dissolution of the resin compsn. into the organic solvent is effective in high compsn. uniformity, optical isotropy and heat resistance.(0/0)

IW - OPTICAL ELEMENT MANUFACTURE RESIN COMPOSITION AROMATIC POLYVINYL POLYMER POLYPHENYLENE ETHER

IKW - OPTICAL ELEMENT MANUFACTURE RESIN COMPOSITION AROMATIC POLYVINYL POLYMER POLYPHENYLENE ETHER

NC - 001

OPD - 1987-04-14

ORD - 1988-10-24

PAW - (SUMO) SUMITOMO CHEM IND KK

TI - Optical element mfr. - using resin compsn. of aromatic vinyl] polymer and polyphenylene ether

PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

To: <div style="text-align: center;">see form PCT/ISA/220</div>	<div style="text-align: center;"> RECEIVED NOV 15 2004 IPO GENERAL ELECTRIC </div> <div style="text-align: center;"> PCT CO-WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY (PCT Rule 43bis.1) </div>
Date of mailing (day/month/year) see form PCT/ISA/210 (second sheet)	
Applicant's or agent's file reference see form PCT/ISA/220 126750	FOR FURTHER ACTION See paragraph 2 below 11 NOV 2004
International application No. PCT/US2004/027237	International filing date (day/month/year) 23.08.2004
Priority date (day/month/year) 26.08.2003	
International Patent Classification (IPC) or both national classification and IPC C08L71/00, C08L25/06, D01D1/10, B29D17/00, B29K71/00	
Applicant GENERAL ELECTRIC COMPANY	

1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☒ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the international application
- ☐ Box No. VIII Certain observations on the international application

2. **FURTHER ACTION**

If a demand for international preliminary examination is made, this opinion will usually be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA"). However, this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1bis(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of three months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.


For further options, see Form PCT/ISA/220.

3. For further details, see notes to Form PCT/ISA/220.

ACTION DUE 6-26-2005

ATTORNEY CJW

LOGGED BY lc 11-17-04

Name and mailing address of the ISA: <div style="text-align: center;">  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465 </div>	Authorized Officer Frison, C Telephone No. +49 89 2399-8519
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6-26-05

**WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY**

International application No.
PCT/US2004/027237

Box No. I Basis of the opinion

1. With regard to the **language**, this opinion has been established on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ This opinion has been established on the basis of a translation from the original language into the following language , which is the language of a translation furnished for the purposes of international search (under Rules 12.3 and 23.1(b)).
2. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application and necessary to the claimed invention, this opinion has been established on the basis of:
 - a. type of material:

☐ a sequence listing

☐ table(s) related to the sequence listing
 - b. format of material:

☐ in written format

☐ in computer readable form
 - c. time of filing/furnishing:

☐ contained in the international application as filed.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority for the purposes of search.
3. ☐ In addition, in the case that more than one version or copy of a sequence listing and/or table relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
4. Additional comments:

**WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY**

International application No.
PCT/US2004/027237

Box No. II Priority

1. ☒ The following document has not been furnished:

☒ copy of the earlier application whose priority has been claimed (Rule 43*bis*.1 and 66.7(a)).

☐ translation of the earlier application whose priority has been claimed (Rule 43*bis*.1 and 66.7(b)).

Consequently it has not been possible to consider the validity of the priority claim. This opinion has nevertheless been established on the assumption that the relevant date is the claimed priority date.

2. ☐ This opinion has been established as if no priority had been claimed due to the fact that the priority claim has been found invalid (Rules 43*bis*.1 and 64.1). Thus for the purposes of this opinion, the international filing date indicated above is considered to be the relevant date.

3. Additional observations, if necessary:

Box No. V Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	
	No: Claims	1-11
Inventive step (IS)	Yes: Claims	
	No: Claims	1-11
Industrial applicability (IA)	Yes: Claims	1-11
	No: Claims	

2. Citations and explanations

see separate sheet

Re Item V.

1 The following document is referred to in this communication:

D1 : DATABASE WPI Section Ch, Week 198848 Derwent Publications Ltd.,
London, GB; Class A14, AN 1988-342863 XP002303242 & JP 63 256427 A
(SUMITOMO CHEM IND KK) 24 October 1988 (1988-10-24)

2. Art. 33(2) PCT

Claim 1

Document D1 recites an optical material based on a composition of an aromatic vinyl polymer and polyphenylene ether which has been filtered to remove fine particulate foreign matters. Document D1 therefore discloses all the technical features of the independent claim 1 of the application. Since D1 was only provided as an abstract to the Examining Division it can not be decided up to now whether D1 anticipates the subject-matter claimed or not. In order to allow a final assessment and to streamline the proceedings the applicant is kindly asked to provide a translation of D1. Document D1 probably does not inform the reader about the size of particulates remaining in the composition. The applicant is kindly requested to show to the Examining Division that any example disclosed in D1 does not satisfy the requirements disclosed in claim 1.

At present the subject-matter of claim 1 can not be considered as being novel.

Claim 3

The process of claim 3 is expressed in terms of results to be achieved, without explicitly informing about the essential features required in order to achieve said result. Claim 3 is thus not clear. Besides, melt-filtering techniques are already used to purify polymers (see US 4 500 706).

Claims 8, 9

The articles of claims 8, 9 are known from document D1.